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(54) FILM-FORMING COMPOSITION, FORMATION OF FILM AND LOW- DENSITY FILM

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a film-forming composition capable of imparting a low density film excellent in dielectric constant characteristics and water absorbency characteristics and useful as an interlayer insulating film in semiconductor elements, etc., and being an uniform coating film, and also excellent in storage stability.

SOLUTION: This film-forming composition comprises (A) silane compounds composed of $R_2R_3Si(OR_1)_2$ and/or $R_2Si(OR_1)_3$, and $Si(OR_1)_4$ (wherein R_1 to R_3 are each a monovalent organic group) or its hydrolyzate and/or condensation product, (B) a polyether represented by the general formula: PEO_n-R (wherein PEO is a polyethylene oxide unit; R is a 5-30C monovalent organic group; n is an integer of 5-50) and (C) at least one kind selected from the group consisting of an alcohol-based solvent, a ketone-based solvent, an amide-based solvent and an ester-based solvent.

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Notes:

1. Untranslatable words are replaced with asterisks (****).
2. Texts in the figures are not translated and shown as it is.

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Dictionary: Last updated 03/24/2006 / Priority: 1. Chemistry / 2. Natural sciences / 3. Technical term

CLAIMS

[Claim(s)]

[Claim 1] (A) (A-1) The compound expressed with the compound expressed with a following general formula (1), and/or (A-2) a following general formula (2), (A-3) The silane compound containing the compound expressed with a following general formula (3), its hydrolyzate and/or its condensate, $R_2 R_3 Si (OR_1)_2$ (1)

$R_2 Si(OR_1)_3$ (2)

$Si(OR_1)_4$ (3)

(Even if R_1 - R_3 is the same, it may differ, and it shows a univalent organic group, respectively.)

(B) general formula $PEOn-R$ (however, PEO -- a polyethylene oxide unit --) R -- the univalent organic group of a carbon number 5-30, and n -- the integer of 5-50 -- being shown -- the constituent for film formation characterized by containing at least one sort of solvents chosen from the group of the polyether expressed and (C) alcoholic solvent, ketone solvent, an amide system solvent, and ester solvent.

[Claim 2] When each component is converted into a complete hydrolysis condensate among a component, a component (A-3) (A) A component (A-1), (A-2) The constituent for film formation according to claim 1 which is 5 to 60 weight % to the total amount of a component and (A-3) a component, and is weight $[(A-1) \text{ component}] < [(A-2) \text{ weight of a component}]$.

[Claim 3] (C) The constituent for film formation according to claim 1 whose solvents are alcoholic solvent and/or ketone solvent.

[Claim 4] The formation method of the film characterized by applying Claim 1 - the constituent for film formation given in 3 any 1 clauses to a substrate, and heating them.

[Claim 5] The formation method of the film according to claim 4 which applies Claim 1 - the constituent for film formation given in 3 any 1 clauses to a substrate, heats at the temperature of under the decomposition temperature of the (B) component, is made to harden a part of (A)

component, and is subsequently heated and stiffened at the temperature more than the decomposition temperature of the above-mentioned (B) component.

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[Claim 6] The formation method of the film according to claim 4 which applies Claim 1 - the constituent for film formation given in 3 any 1 clauses to a substrate, and is heated and stiffened at the temperature more than the decomposition temperature of the (B) component.

[Claim 7] The low density-ized film obtained by the formation method of a film Claim 4 - given in 6 any 1 clauses.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] About the constituent for film formation, in more detail, this invention can form the paint film which was excellent in permittivity characteristics and water absorption characteristics, and was excellent in paint film homogeneity as an interlayer insulation film material in a semiconductor device etc., and relates to the constituent for film formation which is moreover excellent also in storage stability.

[0002]

[Description of the Prior Art] The silica (SiO₂) film formed in vacuum processes, such as a CVD method, is conventionally used abundantly as an interlayer insulation film in a semiconductor device etc. And the applied type insulating film which uses as a principal component the hydrolysis product of the tetra-alkoxy run called a SOG (Spin on Glass) film for the purpose of forming a recent years more uniform interlayer insulation film is also used. Moreover, the interlayer insulation film of low permittivity which uses polyorganosiloxane called Organicity SOG as a principal component is developed with high integration of a semiconductor device etc. However, the electric insulation between the more excellent conductors is demanded with the further high integration and the further multilayering of a semiconductor device etc., therefore the interlayer insulation film material which is low permittivity more and is excellent in cracking resistance is called for.

[0003] So, the spreading setup-of-tooling product for insulating film formation of low permittivity is indicated by JP,H6-181201,A as an interlayer insulation film material. This spreading setup-of-tooling product has low absorptivity, and aims at offering the insulating film of the semiconductor device excellent in crack-proof nature. The organometallic compound with which the composition contains at least one sort of elements chosen from titanium, zirconium, niobium, and a tantalum, The number average molecular weight which comes to carry out condensation polymerization of the organic silicon compound which has at least one alkoxy group to intramolecular is the spreading setup-of-tooling product for insulating film formation which uses 500 or more oligomer as a principal component. However, the permittivity of the

conventional inorganic system interlayer insulation film material is 3.0 or more, and is inadequate for high integration.

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[0004]

[Problem(s) to be Solved by the Invention] This invention aims at offering the constituent for film formation which can form the paint film excellent in thickness homogeneity as an interlayer insulation film in a semiconductor device etc. in more detail about the constituent for film formation for solving the above-mentioned problem, is excellent in permittivity characteristics and water absorption characteristics, and is moreover excellent also in storage stability.

[0005]

[Means for Solving the Problem] The compound expressed with the compound (henceforth "a compound (1)") by which this invention is expressed with the (A) (A-1) following general formula (1), and/or (A-2) a following general formula (2) (henceforth "a compound (2)"), (A-3) The compound expressed with a following general formula (3) (henceforth "a compound (3)"),

The silane compound containing **, its hydrolyzate and/or its condensate (respectively henceforth "a component (A-1)", "a component (A-2)", and "a component (A-3)"), $R_2 R_3 Si(OR_1)_2$ (1)

$R_2 Si(OR_1)_3$ (2)

$Si(OR_1)_4$ (3)

(Even if $R_1 - R_3$ is the same, it may differ, and it shows a univalent organic group, respectively.)

(B) general formula PEO_n-R (however, PEO -- a polyethylene oxide unit --) R -- the univalent organic group of a carbon number 5-30, and n -- the integer of 5-50 -- being shown -- it is related with the constituent for film formation characterized by containing at least one sort of solvents chosen from the group of the polyether expressed and (C) alcoholic solvent, ketone solvent, an amide system solvent, and ester solvent. Here, among the (A) component, when each component is converted into a complete hydrolysis condensate, it is desirable that a component (A-3) is 5 to 60 weight % to the total amount of a component (A-1), a component (A-2), and (A-3) a component, and it is weight [% of [(A-1) component] < [(A-2) weight of a component]]. Moreover, as a (C) solvent, when alcoholic solvent and/or ketone solvent are used, it excels in the storage stability of the constituent obtained. Next, this invention relates to the formation method of the film characterized by applying the above-mentioned constituent for film formation to a substrate, and heating it. As the formation method of the above-mentioned film, the constituent for the ** above-mentioned film formation is applied to a substrate here. (B) It is desirable to heat at the temperature of under the decomposition temperature of a component, to stiffen a part of (A) component, to make it heat and harden at the temperature more than the decomposition temperature of the above-mentioned (B) component subsequently, or to apply the constituent for the ** above-mentioned film formation to a

substrate, and to make it heat and harden at the temperature more than the decomposition temperature of the (B) component. Next, this invention relates to the low density-ized film obtained by the formation method of the above-mentioned film.

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[0006]

[Embodiment of the Invention] The (C) solvent is used for this invention as a solvent of (A) - (B) component as base polymer which forms a film using specific (B) polyether as a material which forms porosity for the (A) component [compound (1) - (3), its hydrolyzate, and/or its condensate] again. In this way, if the constituent of this invention containing (A) - (C) component is excellent in storage stability and this constituent is applied to base materials, such as a silicon wafer, with immersion or a spin coat method, it can fully fill the slot between minute patterns, for example. And if removal of (B) polyether and the (C) solvent and the heat polycondensation of the (A) component are performed with heating, while the (A) component will form the film of glassiness or a huge polymer, micropore is formed by decomposing and removing (B) polyether. It excels in paint film homogeneity, the film obtained is a porous low density film, its permittivity is low, since it is low absorptivity, it is excellent in the stability of an electrical property, and it can form interlayer insulation film material.

[0007] Here, the above-mentioned hydrolyzates in the (A) component may be that into which all the R1 O-groups contained in the above-mentioned (A) component do not need to be hydrolyzed into, for example, only one piece is hydrolyzed, the things into which two or more pieces are hydrolyzed, or these mixtures. Moreover, although the silanol group of the hydrolyzate of the (A) component condenses the above-mentioned condensate in the (A) component and Si-O-Si binding is formed Although what all silanol groups do not need to condense and some [few] silanol groups condensed in this invention differs from the grade of condensation, it is the concept which included the mixture etc.

[0008] (A) - (C) component used for this invention is explained hereafter, and, subsequently the preparation methods of the constituent of this invention etc. are explained in full detail.

[0009] (A) a component (A-3) is used for a component (A) component as an essential ingredient -- this -- and/or (A-1) (A-2), use together. The above-mentioned general formula (1) In - (3), an alkyl group, an aryl group, an allyl group, a glycidyl group, etc. can be mentioned as a univalent organic group. Here, as an alkyl group, a methyl group, an ethyl group, a propyl group, butyl, etc. are mentioned, it is a carbon number 1-5 preferably, and even if these alkyl groups are catenoid, you may branch and the hydrogen atom may be further replaced by the fluorine atom etc. General formula (1) In - (3), a phenyl group, a naphthyl group, a methylphenyl machine, an ethyl phenyl group, a chlorophenyl machine, a bromo phenyl group, a fluoro phenyl group, etc. can be mentioned as an aryl group.

[0010] As an example of a compound expressed with a general formula (1)

Dimethyldimethoxysilane, dimethyldiethoxysilane, dimethyl G n-propoxysilane, Dimethyl G iso-

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propoxysilane, dimethyl G n-butoxysilane, Dimethyl G sec-butoxysilane, dimethyl G tert-butoxysilane, JIMECHIRUJI phenoxy silane, diethyldimethoxysilane, diethyldiethoxysilane, Diethyl G n-propoxysilane, diethyl G iso-propoxysilane, Diethyl G n-butoxysilane, diethyl G sec-butoxysilane, Diethyl G tert-butoxysilane, JIECHIRUJI phenoxy silane, G n-propyl dimethoxysilane, di-n-propyl diethoxysilane, G n-propyl G n-propoxysilane, G n-propyl G iso-propoxysilane, G n-propyl G n-butoxysilane, G n-propyl G sec-butoxysilane, G n-propyl G tert-butoxysilane, G n-propyl G phenoxy silane, G iso-propyl dimethoxysilane, G iso-propyl diethoxysilane, G iso-propyl G n-propoxysilane, G iso-propyl G iso-propoxysilane, G iso-propyl G n-butoxysilane, G iso-propyl G sec-butoxysilane, G iso-propyl G tert-butoxysilane, G iso-propyl G phenoxy silane, Di-n-butyl dimethoxysilane, di-n-butyl diethoxysilane, G n-butyl G n-propoxysilane, G n-butyl G iso-propoxysilane, G n-butyl G n-butoxysilane, G n-butyl G sec-butoxysilane, G n-butyl G tert-butoxysilane, G n-butyl G phenoxy silane, G sec-butyl dimethoxysilane, G sec-butyl diethoxysilane, G sec-butyl G n-propoxysilane, G sec-butyl G iso-propoxysilane, G sec-butyl G n-butoxysilane, G sec-butyl G sec-butoxysilane, G sec-butyl G tert-butoxysilane, G sec-butyl G phenoxy silane, G tert-butyl dimethoxysilane, G tert-butyl diethoxysilane, G tert-butyl G n-propoxysilane, G tert-butyl G iso-propoxysilane, G tert-butyl G n-butoxysilane, G tert-butyl G sec-butoxysilane, G tert-butyl G tert-butoxysilane, G tert-butyl G phenoxy silane, Diphenyldimethoxysilane, diphenyl G ethoxy silane, diphenyl G n-propoxysilane, Diphenyl G iso-propoxysilane, diphenyl G n-butoxysilane, diphenyl G sec-butoxysilane, diphenyl G tert-butoxysilane, a diphenyl JIFENOKI gardenia fruit run, etc. are mentioned.

[0011] moreover, as an example of a compound expressed with a general formula (2) Methyl trimethoxysilane, methyltriethoxysilane, methyl tree n-propoxysilane, Methyl tree iso-propoxysilane, methyl tree n-butoxysilane, Methyl tree sec-butoxysilane, methyl tree tert-butoxysilane, Methyl triphenoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, Ethyl tree n-propoxysilane, ethyl tree iso-propoxysilane, Ethyl tree n-butoxysilane, ethyl tree sec-butoxysilane, Ethyl tree tert-butoxysilane, ethyl triphenoxysilane, n-propyltrimethoxysilane, n-propyl triethoxysilane, n-propyl tree n-propoxysilane, n-propyl tree iso-propoxysilane, n-propyl tree n-butoxysilane, n-propyl tree sec-butoxysilane, n-propyl tree tert-butoxysilane, n-propyl triphenoxysilane, i-propyltrimethoxysilane, i-propyl triethoxysilane, i-propyl tree n-propoxysilane, i-propyl tree iso-propoxysilane, i-propyl tree n-butoxysilane, i-propyl tree sec-butoxysilane, i-propyl tree tert-butoxysilane, i-propyl triphenoxysilane, n-butyltrimethoxysilane, N-butyltriethoxysilane, n-butyl tree n-propoxysilane, N-butyl tree iso-propoxysilane, n-butyl tree n-butoxysilane, N-butyl tree sec-butoxysilane, n-butyl tree tert-butoxysilane, N-butyl triphenoxysilane, sec-butyltrimethoxysilane, sec-butyl triethoxysilane, sec-butyl tree n-propoxysilane, sec-butyl tree iso-propoxysilane, sec-butyl tree n-butoxysilane, sec-butyl tree sec-butoxysilane, sec-butyl tree tert-butoxysilane, sec-butyl triphenoxysilane, t-

butyltrimethoxysilane, t-butyltriethoxysilane, t-butyl tree n-propoxysilane, t-butyl tree iso-propoxysilane, t-butyl tree n-butoxysilane, t-butyl tree sec-butoxysilane, t-butyl tree tert-butoxysilane, t-butyl triphenoxysilane, Phenyltrimethoxysilane, phenyltriethoxysilane, phenyl tree n-propoxysilane, Phenyl tree iso-propoxysilane, phenyl tree n-butoxysilane, Phenyl tree sec-butoxysilane, phenyl tree tert-butoxysilane, Others, vinyltrimetoxysilane which are phenyl triphenoxysilane etc., Vinyltriethoxysilane, gamma-aminopropyl trimethoxysilane, gamma-aminopropyl triethoxysilane, gamma-glycidoxypropyltrimetoxysilane, gamma-glycidoxy propyltriethoxysilane, gamma-bird FURORO propyltrimethoxysilane, gamma-bird FURORO propyl triethoxysilane, etc. are mentioned.

[0012] furthermore, as an example of a compound expressed with a general formula (3) Tetramethoxy silane, tetra-ethoxy silane, tetra-n-propoxysilane, tetra iso-propoxysilane, a tetra-n-butoxy run, tetra sec-butoxysilane, tetra tert-butoxysilane, tetra-phenoxy silane, etc. are mentioned.

[0013] Compound (1) Among - (3), preferably Tetramethoxy silane, Tetra-ethoxy silane, tetra-n-propoxysilane, tetra iso-propoxysilane, Methyl trimetoxysilane, methyltriethoxysilane, methyl tree n-propoxysilane, Methyl tree iso-propoxysilane, phenyltrimethoxysilane, Are phenyltriethoxysilane, dimethyldimethoxysilane, dimethyldiethoxysilane, diphenyldimethoxysilane, and diphenyl diethoxysilane, and [especially] preferably They are tetra-METOKI xylan, tetra-ethoxy silane, methyl trimetoxysilane, methyltriethoxysilane, dimethyldimethoxysilane, and dimethyldiethoxysilane. these -- one sort -- or you may use two or more sorts simultaneously.

[0014] When making compound (1) - (3) which constitutes the above-mentioned (A) component hydrolyze and condense, it is desirable to use 0.25-3mol [per / which is expressed with R1 O- / 1mol of groups] water, and especially the thing for which the water which is 0.3-2.5mol is added is desirable. If it is a value within the limits whose quantity of the water to add is 0.25-3mol, there is no possibility that the homogeneity of a paint film may fall, and it is because there are little deposit of the polymer under a hydrolysis and condensation reaction and fear of gelling.

[0015] (A) When making compound (1) - (3) which constitutes a component hydrolyze and condense, you may use a catalyst. In this case, as a catalyst to be used, metal chelate compound, an organic acid, an inorganic acid, an organic base, and an inorganic base can be mentioned.

[0016] As metal chelate compound, for example Bird ethoxy mono-(acetylacetonato) titanium, Tree n-propoxy mono-(acetylacetonato) titanium, tree i-propoxy mono-(acetylacetonato) titanium, Tree n-butoxy mono-(acetylacetonato) titanium, tree sec-butoxy mono-(acetylacetonato) titanium, Tree t-butoxy mono-(acetylacetonato) titanium, diethoxy bis (acetylacetonato) titanium, G n-propoxy bis(acetylacetonato) titanium, G i-propoxy bis

(acetylacetonato) titanium, G n-butoxy bis(acetylacetonato) titanium, G sec-butoxy bis(acetylacetonato) titanium, G t-butoxy bis(acetylacetonato) titanium, mono-ethoxy tris(acetylacetonato) titanium, Monod n-propoxy tris(acetylacetonato) titanium, Monod i-propoxy tris(acetylacetonato) titanium, Monod n-butoxy tris(acetylacetonato) titanium, Monod sec-butoxy tris(acetylacetonato) titanium, Monod t-butoxy tris(acetylacetonato) titanium, tetrakis(acetylacetonato) titanium, Bird ethoxy mono-(ethyl acetoacetate) titanium, tree n-propoxy mono-(ethyl acetoacetate) titanium, Tree i-propoxy mono-(ethyl acetoacetate) titanium, tree n-butoxy mono-(ethyl acetoacetate) titanium, Tree sec-butoxy mono-(ethyl acetoacetate) titanium, Tree t-butoxy mono-(ethyl acetoacetate) titanium, diethoxy bis(ethyl acetoacetate) titanium, G n-propoxy bis(ethyl acetoacetate) titanium, G i-propoxy bis(ethyl acetoacetate) titanium, G n-butoxy bis(ethyl acetoacetate) titanium, G sec-butoxy bis(ethyl acetoacetate) titanium, G t-butoxy bis(ethyl acetoacetate) titanium, mono-ethoxy tris(ethyl acetoacetate) titanium, Monod n-propoxy tris(ethyl acetoacetate) titanium, Monod i-propoxy tris(ethyl acetoacetate) titanium, Monod n-butoxy tris(ethyl acetoacetate) titanium, Monod sec-butoxy tris(ethyl acetoacetate) titanium, Monod t-butoxy tris(ethyl acetoacetate) titanium, tetrakis(ethyl acetoacetate) titanium, Titanium chelate compound, such as mono-(acetylacetonato) tris(ethyl acetoacetate) titanium, bis(acetylacetonato) bis(ethyl acetoacetate) titanium, and tris(acetylacetonato) mono-(ethyl acetoacetate) titanium;

[0017] Bird ethoxy mono-(acetylacetonato) zirconium, tree n-propoxy mono-(acetylacetonato) zirconium, Tree i-propoxy mono-(acetylacetonato) zirconium, Tree n-butoxy mono-(acetylacetonato) zirconium, Tree sec-butoxy mono-(acetylacetonato) zirconium, Tree t-butoxy mono-(acetylacetonato) zirconium, Diethoxy bis(acetylacetonato) zirconium, G n-propoxy bis(acetylacetonato) zirconium, G i-propoxy bis(acetylacetonato) zirconium, G n-butoxy bis(acetylacetonato) zirconium, G sec-butoxy bis(acetylacetonato) zirconium, G t-butoxy bis(acetylacetonato) zirconium, mono-ethoxy tris(acetylacetonato) zirconium, Monod n-propoxy tris(acetylacetonato) zirconium, Monod i-propoxy tris(acetylacetonato) zirconium, Monod n-butoxy tris(acetylacetonato) zirconium, Monod sec-butoxy tris(acetylacetonato) zirconium, Monod t-butoxy tris(acetylacetonato) zirconium, Tetrakis(acetylacetonato) zirconium, bird ethoxy mono-(ethyl acetoacetate) zirconium, Tree n-propoxy mono-(ethyl acetoacetate) zirconium, Tree i-propoxy mono-(ethyl acetoacetate) zirconium, Tree n-butoxy mono-(ethyl acetoacetate) zirconium, Tree sec-butoxy mono-(ethyl acetoacetate) zirconium, Tree t-butoxy mono-(ethyl acetoacetate) zirconium, Diethoxy bis(ethyl acetoacetate) zirconium, G n-propoxy bis(ethyl acetoacetate) zirconium, G i-propoxy bis(ethyl acetoacetate) zirconium, G n-butoxy bis(ethyl acetoacetate) zirconium, G sec-butoxy bis(ethyl acetoacetate) zirconium, G t-butoxy bis(ethyl acetoacetate) zirconium, mono-ethoxy tris(ethyl acetoacetate) zirconium Monod n-propoxy tris(ethyl acetoacetate) zirconium, Monod i-propoxy tris(ethyl acetoacetate) zirconium, Monod n-butoxy tris(ethyl acetoacetate) zirconium, Monod sec-butoxy tris(ethyl

acetoacetate) zirconium, Monod t-butoxy tris (ethyl acetoacetate) zirconium, Tetrakis (ethyl acetoacetate) zirconium, mono-(acetylacetonato) tris (ethyl acetoacetate) zirconium, Zirconium^{8/20} chelate compound, such as bis(acetylacetonato) bis(ethyl acetoacetate) zirconium and tris (acetylacetonato) mono-(ethyl acetoacetate) zirconium;

[0018] Aluminum chelate compound [, such as a tris (acetylacetonato) aluminium and a tris (ethyl acetoacetate) aluminium,]; etc. can be mentioned.

[0019] As an organic acid, for example Acetic acid, propionic acid, butanoic acid, pentanoic acid, Hexanoic acid, oenanthic acid, octanoic acid, nonoic acid, decanoic acid, oxalic acid, Maleic acid, methylmalonic acid, adipic acid, sebacic acid, gallic acid, Butanoic acid, mellitic acid, arachidonic acid, Illicium religiosum, 2-ethyl hexanoic acid, Oleic acid, stearin acid, linolic acid, the Reno Laing acid, salicylic acid, Benzoic acid, p-aminobenzoic acid, p-toluenesulfonic acid, benzenesulfonic acid, monochloroacetic acid, dichloroacetic acid, trichloroacetic acid, trifluoroacetic acid, formic acid, malonic acid, sulfonic acid, phthalic acid, fumaric acid, citric acid, tartaric acid, etc. can be mentioned. As an inorganic acid, hydrochloric acid, nitric acid, sulfuric acid, fluoric acid, phosphoric acid, etc. can be mentioned, for example.

[0020] As an organic base, for example Pyridine, pyrrole, piperazine, pyrrolidine, Piperidine, picoline, trimethylamine, triethylamine, monoethanolamine, Diethanolamine, dimethyl monoethanolamine, monomethyl diethanolamine, triethanolamine, a diazabicyclo clan, diazabicyclo nonane, diazabicycloundecen, tetramethylammonium hydroxide, etc. can be mentioned. As an inorganic base, ammonia, sodium hydroxide, potassium hydroxide, barium hydroxide, a calcium hydroxide, etc. can be mentioned, for example.

[0021] Metal chelate compound, an organic acid, and an inorganic acid are desirable among these catalysts, and an organic acid can be mentioned more preferably. Especially as an organic acid, acetic acid, oxalic acid, maleic acid, and malonic acid are desirable. As a catalyst, when an organic acid is used, a deposit of the polymer under a hydrolysis and condensation reaction and fear of gelling are desirable few. these catalysts -- one sort -- or you may use two or more sorts simultaneously.

[0022] 0.00001-0.05mol of the amount of the above-mentioned catalyst used is usually 0.00001-0.01mol preferably to 1mol of total amounts of compound (1) - (3).

[0023] (A) when a component is the condensate of compound (1) - (3), the molecular weight is a weight average molecular weight of polystyrene conversion -- usually -- 500-300,000 -- desirable -- 700-200,000 -- it is 1,000 to about 100,000 still more preferably.

[0024] When [moreover,] each component is converted into a complete hydrolysis condensate among the (A) component (A-3) A component is 5 to 40 weight % still more preferably five to 50weight % preferably five to 60weight % to the total amount of a component (A-1), a component (A-2), and (A-3) a component, and it is weight][of [(A-1) component] < [(A-2) weight of a component]. At a rate of each component converted into the complete

hydrolysis condensate, the mechanical strength of the paint film with which a component (A-3) is obtained in less than 5weight % of the total amount of - (A-1) (A-3) component falls, on the other hand, if it exceeds 60 weight %, absorptivity will become high and an electrical property will fall. Moreover, the hardness of the paint film obtained as the weight of a component (A-1) is more than the weight of a component (A-2) is inferior. In addition, in this invention, a complete hydrolysis condensate is SiOR1 of compound (1) - (3). What the machine hydrolyzed 100% and became a SiOH group, condensed still more completely and became siloxane structure is said.

[0025] (B) As polyether (B) polyether, it is the polyoxyethylene system polymer expressed with a following general formula (4).

PEOn-R (4)

(However, PEO shows a polyethylene oxide unit, R shows the univalent organic group of a carbon number 5-30, and n shows the integer of 5-50.)

[0026] The still more detailed structure of this (B) polyether is expressed with a following general formula (5).

R4 -(CH2 CH2 O)n-R5 (5)

R4 expresses hydroxyl, a carboxyl group, an alkoxy group, a bird alkoxy silyl machine, and alkyl group (bird alkoxy silyl) ** among [type, in this case, the carbon number of an alkoxy group is five or less, R5 expresses the univalent organic group of a carbon number 5-30, and n shows the integer of 5-50.]

[0027] In the above-mentioned general formula (5), it is R4. if it carries out Hydroxyl, a carboxyl group methoxy group, an ethoxy group, a propoxy group, A trimethoxy siloxy machine, a bird ethoxy siloxy machine, a tripropoxy siloxy machine, A trimethoxysilyl methoxy group, a triethoxy silyl methoxy group, 2-trimethoxysilyl ethoxy group, 2-triethoxy silyl ethoxy group, 3-trimethoxysilyl propoxy group, 3-triethoxy silyl propoxy group, etc. can be mentioned, hydroxyl and a methoxy group can be mentioned preferably, and hydroxyl can be mentioned especially preferably. Moreover, R5 which is a univalent organic group in the above-mentioned general formula (5) If it carries out, an alkyl group, an alkylphenyl machine, an alkoxy carbonyl group, etc. can be mentioned. Here, as an alkyl group, the alkyl group of a carbon number 5-30 can be mentioned, and a normal chain alkyl group can be mentioned preferably. As an alkylphenyl machine, the phenyl group which has the alkyl group of a carbon number 1-24 can be mentioned, and the alkyl group which has a normal chain alkyl group in para position preferably can be mentioned. As an alkoxy carbonyl group, the alkoxy carbonyl group of a carbon number 5-30 can be mentioned, and a normal chain alkoxy carbonyl group can be mentioned preferably.

[0028] As an example of (B) polyether expressed with a general formula (5) Polyethylene glycol mono-pentyl ether, polyethylene glycol mono-hexyl ether, Polyethylene glycol

MONOPEPU chill ether, poly ethylene glycol monooctyl ether, Polyethylene glycol mono-nonyl ether, polyethylene glycol MONODEKA nil ether, Polyethylene glycol mono-undecanyl ether, polyethylene glycol mono-dodecanyl ether, Polyethylene glycol MONOTORIDEKA NIRUETERU, polyethylene glycol MONOTETORADEKA nil ether, Polyethylene glycol MONOPENTADEKA nil ether, polyethylene glycol MONOHEKISADEKA nil ether, Polyethylene glycol mono-heptadeca NIRUETERU, polyethylene glycol mono-octadecanyl ether, Polyethylene glycol MONONONADEKA NIRUETERU, polyethylene glycol mono-icosanyl ether, Polyethylene glycol MONOHENIKOSA nil ether, polyethylene glycol MONODOKOSA nil ether, polyethylene glycol MONOTORIKOSA nil ether, polyethylene glycol mono-tetracosa NIRUETERU, Polyethylene glycol MONOPENTAKOSA nil ether, polyethylene glycol mono-hexacosa NIRUETERU, Polyethylene glycol MONOHEPUTAKOSA nil ether, polyethylene glycol MONOOKUTAKOSA nil ether, Polyethylene glycol alkyl ether and methyl ether of those, such as polyethylene glycol MONONONAKOSA NIRUETERU and polyethylene glycol MONOTORIAKONTA nil ether, Ethyl ether, propyl ether, trimethoxysilyl ether, Triethoxy silyl ether, tripropoxy silyl ether, trimethoxysilyl methyl ether, Polyethylene glycol alkyl ether derivatives, such as triethoxy silyl methyl ether, 2-trimethoxysilyl ethyl ether, 2-triethoxy silyl ethyl ether, 3-trimethoxysilylpropyl ether, and 3-triethoxy silyl propyl ether;

[0029] Polyethylene glycol ****- p-methylphenyl ether, polyethylene glycol ****- p-ethyl phenyl ether, Polyethylene glycol ****- p-propyl phenyl ether, polyethylene glycol ****- p-butyl phenyl ether, Polyethylene glycol ****- p-pentyl phenyl ether, polyethylene glycol ****- p-hexyl phenyl ether, Polyethylene glycol ****- p-PEPUCHIRU phenyl ether, polyethylene glycol ****- p-octyl phenyl ether, Polyethylene glycol ****- p-nonyl phenyl ether, polyethylene glycol ****- p-deca nil phenyl ether, Polyethylene glycol ****- p-undecanyl phenyl ether, Polyethylene glycol ****- p-dodecanyl phenyl ether, polyethylene glycol ****- p-trideca nil phenyl ether, Polyethylene glycol ****- p-tetra-deca nil phenyl ether, polyethylene glycol ****- p-penta deca nil phenyl ether, polyethylene glycol ****- p-hexa deca nil phenyl ether, Polyethylene glycol ****- p-heptadeca nil phenyl ether, Polyethylene glycol ****- p-octadecanyl phenyl ether, Polyethylene glycol ****- p-nonadeca nil phenyl ether, Polyethylene glycol ****- p-icosanyl phenyl ether, polyethylene glycol ****- p-HENIKOSA nil phenyl ether, Polyethylene glycol ****- p-docosa nil phenyl ether, polyethylene glycol ****- p-TORIKOSA nil phenyl ether, Polyethylene-glycols p-alkylphenyl ether and its methyl ether, such as polyethylene glycol ****- p-tetracosa nil phenyl ether, Ethyl ether, propyl ether, trimethoxysilyl ether, Triethoxy silyl ether, tripropoxy silyl ether, trimethoxysilyl methyl ether, Triethoxy silyl methyl ether, 2-trimethoxysilyl ethyl ether, Polyethylene-glycols p-alkylphenyl ether derivatives, such as 2-triethoxy silyl ethyl ether, 3-trimethoxysilylpropyl ether, and 3-triethoxy silyl propyl ether;

[0030] Polyethylene glycol mono-pentanoic acid ester, polyethylene glycol mono-hexanoate, Polyethylene glycol mono-oenanthic acid ester, polyethylene glycol mono-octoate,

Polyethylene glycol mono-nonoic acid ester, polyethylene glycol mono-decanoate, Polyethylene glycol mono-undecanoic acid ester, polyethylene glycol mono-dodecanoic acid ester, Polyethylene glycol mono-tridecane acid ester, polyethylene glycol mono-tetradecane acid ester, Polyethylene glycol mono-pentadecane acid ester, polyethylene glycol mono-hexadecanoic acid ester, Polyethylene glycol mono-heptadecanoic acid ester, polyethylene glycol mono-octadecanoic acid ester, Polyethylene glycol mono-nonadecane acid ester, polyethylene glycol mono-icosanoic acid ester, Polyethylene glycol mono-HENIKOSAN acid ester, polyethylene glycol mono-docosanoic acid ester, polyethylene glycol mono-tricosane acid ester, polyethylene glycol mono-tetracosanoic acid ester, polyethylene glycol mono-pentacosane acid ester Polyethylene glycol mono-hexacosanoic acid ester, polyethylene glycol mono-heptacosane acid ester, Polyethylene glycol mono-octacosanoic acid ester, polyethylene glycol mono-nonacosane acid ester, Polyethylene glycol alkyl acid ester and methyl ether of those, such as polyethylene glycol mono-triacontanoic acid ester, Ethyl ether, propyl ether, trimethoxysilyl ether, Triethoxy silyl ether, tripropoxy silyl ether, trimethoxysilyl methyl ether, Triethoxy silyl methyl ether, 2-trimethoxysilyl ethyl ether, Polyethylene glycol alkyl acid ester derivatives, such as 2-triethoxy silyl ethyl ether, 3-trimethoxysilylpropyl ether, and 3-triethoxy silyl propyl ether, etc. can be mentioned.

[0031] In this invention, since the thickness homogeneity of a paint film becomes very good while the density of the paint film obtained will become low and will serve as low dielectricity, if the above-mentioned (B) polyether is used, it is desirable as an interlayer insulation film material between detailed wiring. (B) the weight average molecular weight of polystyrene conversion of polyether -- usually -- 300-3,000 -- it is 300-2,500 preferably.

[0032] (B) the amount of the polyether used receives a (A) component (complete hydrolysis condensate conversion) 100 weight part -- usually -- 1 - 80 weight part -- it is 5 - 65 weight part preferably. Under in 1 weight part, the effect of lowering permittivity is small, and on the other hand, if 80 weight parts are exceeded, mechanical strength will fall.

[0033] (C) The constituent for film formation of solvent this invention dissolves or distributes the (A) component and (B) polyether to at least one sort of solvents chosen from the group of (C) alcoholic solvent, ketone solvent, an amide system solvent, and ester solvent. As alcoholic solvent here Methanol, ethanol, n-propanol, i-propanol, n-butanol, i-butanol, sec-butanol, t-butanol, n-pentanol, i-pentanol, 2-methyl butanol, sec-pentanol, t-pentanol, 3-methoxybutanol, N-hexanol, 2-methyl pentanol, sec-hexanol, 2-ethyl butanol, sec-heptanol, heptanol 3, n-octanol, 2-ethyl hexanol, sec-octanol, n-nonyl alcohol, 2, 6-dimethyl heptanol 4, n-decanol, sec-undecyl alcohol, Bird methyl nonyl alcohol, a sec-tetradecyl alcohol, sec-heptadecyl alcohol, Monoalcohol system solvents, such as phenol, cyclohexanol, methyl cyclohexanol, 3, 3, 5-bird methyl cyclohexanol, benzyl alcohol, and diacetone alcohol;

[0034] Ethylene glycol, 1, 2-propylene glycol, 1,3-butanediol, Pentanediol 2, 4, 2-methyl

pentanediol 2, 4, hexandiol 2, 5, heptane diol 2, 4, 2-ethyl hexanediol 1, 3, diethylene glycol, Polyhydric alcohol system solvents, such as dipropylene glycol, triethylene glycol, and tripropylene glycol; Ethylene glycol monomethyl ether, Ethylene glycol monoethyl ether, ethylene glycol monopropyl ether, Ethylene glycol monobutyl ether, ethylene glycol mono-hexyl ether, Ethylene glycol monophenyl ether, ethylene glycol ****- 2-ethyl butyl ether, Diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, Diethylene glycol monopropyl ether, diethylene glycol monobutyl ether, Diethylene glycol mono-hexyl ether, propylene glycol monomethyl ether, propylene glycol monoethyl ether, propylene glycol monopropyl ether, propylene glycol monobutyl ether Polyhydric alcohol partial ether system solvent [, such as dipropylene glycol monomethyl ether, dipropylene glycol monoethyl ether, and dipropylene glycol monopropyl ether,]; etc. can be mentioned. these alcoholic solvent -- one sort -- or you may use two or more sorts simultaneously.

[0035] The n-propanol among these alcohols, i-propanol, N-butanol, i-butanol, sec-butanol, t-butanol, n-pentanol, i-pentanol, 2-methyl butanol, sec-pentanol, t-pentanol, 3-methoxybutanol, n-hexanol, 2-methyl pentanol, sec-hexanol, 2-ethyl butanol, propylene glycol monomethyl ether, propylene glycol monoethyl ether, propylene glycol monopropyl ether, propylene glycol monobutyl ether, etc. are desirable.

[0036] As ketone solvent, acetone, methyl ethyl ketone, methyl n-propyl ketone, Methyl n-butyl ketone, diethyl ketone, methyl i-butyl ketone, Methyl n-pentyl ketone, ethyl n-butyl ketone, methyl n-hexyl ketone, Non [G i-butyl ketone and bird methyl nona non, / cyclohexanone and 2-hexa], Methylcyclohexanone, 2,4-pentanedione, acetonylacetone, Acetylacetone besides being acetophenone, fenchone, etc., 2, 4-hexane dione, 2, 4-heptane dione, 3, 5-heptane dione, 2, 4-octane dione, 3, 5-octane dione, 2, 4-nonane dione, 3, 5-nonane dione, beta-diketones, such as 5-methyl 2, 4-hexane dione, 2, 2, 6, 6-tetramethyl 3, 5-heptane dione, 1, 1, 1, 5, 5, 5-hexafluoro 2, and 4-heptane dione, are mentioned. these ketone solvent -- one sort -- or you may use two or more sorts simultaneously.

[0037] As an amide system solvent, formamide, N-methyl formamide, N, N dimethylformamide, N-ethyl formamide, N, and N-diethyl formamide, acetamide, N-methyl acetamide, N,N-dimethylacetamide, N-ethyl acetamide, N and N-diethyl acetamide, N-methyl propione amide, N-methyl pyrrolidone, N-formyl morpholine, N-formyl piperidine, N-formyl pyrrolidine, N-acetyl morpholine, N-acetyl piperidine, N-acetyl pyrrolidine, etc. are mentioned. these amide system solvents -- one sort -- or you may use two or more sorts simultaneously.

[0038] As ester solvent, diethyl carbonate, ethylene carbonate, propylene carbonate, Diethyl carbonate, methyl acetate, ethyl acetate, gamma-butyrolactone, gamma-valerolactone, N-propyl acetate, acetic acid i-propyl, acetic acid n-butyl, acetic acid i-butyl, Acetic acid sec-butyl, acetic acid n-pentyl, acetic acid sec-pentyl, acetic acid 3-methoxy butyl, Methyl acetate pentyl, 2-ethylbutyl acetate, 2-ethylhexyl acetate, Benzyl acetate, cyclohexyl acetate,

methylcyclohexyl acetate, n-nonyl acetate, Methyl acetoacetate, ethyl acetoacetate, acetic acid ethylene glycol monomethyl ether, Acetic acid ethylene glycol monoethyl ether, acetic acid diethylene glycol monomethyl ether, Acetic acid diethylene glycol monoethyl ether, acetic acid diethylene glycol ****- n-butyl ether, Acetic acid propylene glycol monomethyl ether, acetic acid propylene glycol monoethyl ether, Acetic acid propylene glycol monopropyl ether, acetic acid propylene glycol monobutyl ether, acetic acid dipropylene glycol monomethyl ether, acetic acid dipropylene glycol monoethyl ether, a JI acetic acid glycol, Acetic acid methoxytriglycol, ethyl propionate, n-butyl propionate, Propionic acid i-amyl, oxalic acid diethyl, oxalic acid di-n-butyl, methyl lactate, ethyl lactate, lactic acid n-butyl, lactic acid n-amyl, diethyl malonate, dimethyl phthalate, diethyl phthalate, etc. are mentioned. these ester solvent -- one sort -- or you may use two or more sorts simultaneously. One sort or two sorts or more can be mixed and used for the above (C) solvent.

[0039] In addition, as a (C) solvent, when alcoholic solvent and/or ketone solvent are used, application properties are good and desirable at the point that the constituent excellent in storage stability is obtained.

[0040] Although the constituent for film formation of this invention contains the above-mentioned (C) solvent, the same solvent can be used for it when it hydrolyzes and/or condenses compound (1) - (3) which constitutes the (A) component.

[0041] The water diluted in water or the (above-mentioned C) solvent in the solvent in which compound (1) - (3) which constitutes the (A) component was specifically dissolved is added intermittently or continuously. Under the present circumstances, you may add beforehand in a solvent, and a catalyst may be underwater dissolved or distributed at the time of water addition. As reaction temperature in this case, 0-100 degrees C is usually 15-90 degrees C preferably.

[0042] In the constituent for film formation obtained by other additive this inventions, you may add components, such as colloidal silica, colloid alumina, and a surface active agent, further. colloidal silica is dispersion liquid which distributed the silicic acid anhydride of the high grade to said hydrophilic organic solvent, for example -- usually -- mean particle diameter -- 5-30 -- 10-20mmicro and solid content concentration are about 10 to 40 weight % preferably m micro. As such colloidal silica, made in Nissan Chemical Industries, Ltd. make, methanol silica sol, and isopropanol silica sol; Catalyst Chemical Industry, Oscar, etc. are mentioned, for example. as colloid alumina -- the alumina sol 520 by Nissan Chemical Industries, Ltd. -- said -- alumina by 100 and said 200; Kawaken Fine Chemicals Co., Ltd. -- clear -- sol and alumina sol 10 -- said -- 132 etc. is mentioned. As a surface active agent, an anion system surface active agent, a cation system surface active agent, an amphoteric surface active agent, etc. are mentioned, and a silicone system surface active agent, a fluorine-containing surface active agent, etc. can be mentioned further, for example.

[0043] It faces preparing the constituent for film formation of preparation method this invention of the constituent for film formation. For example, compound (1) - (3) which constitutes the (A) component is mixed among the (C) solvent, and water is added continuously or intermittently, it hydrolyzes, and after condensing and preparing the (A) component, it is not limited in particular to this that what is necessary is just to add (B) polyether.

[0044] The method of the following ** - ** etc. can be mentioned as an example of the preparation methods of the constituent of this invention.

** (A) the mixture which consists of a compound (1) - (3) and (C) solvent which constitutes a component -- the water of the specified quantity -- in addition, the method of mixing the (B) component, after performing a hydrolysis and a condensation reaction.

** (A) How to mix the (B) component after adding the water of the specified quantity into the mixture which consists of a compound (1) - (3) and (C) component which constitutes a component continuously or intermittently and performing a hydrolysis and a condensation reaction into it.

** (A) How to add the water of the specified quantity to the mixture which consists of the compound (1) - (3) and (B) component and the (C) component which constitute a component, and to perform a hydrolysis and a condensation reaction.

** (A) How to add the water of the specified quantity into the mixture which consists of the compound (1) - (3) and (B) component and the (C) component which constitute a component continuously or intermittently, and to perform a hydrolysis and a condensation reaction into it.

[0045] Thus, preferably, the total-solids concentration of the constituent of this invention obtained is 2 to 30 weight %, and is suitably adjusted according to the purpose of use. The thickness of a paint film becomes that the total-solids concentration of a constituent is 2 to 30 weight % with the suitable range, and preservation stability is also more excellent. In addition, if this total-solids concentration needs to be adjusted, it will be performed by dilution by concentration or the above-mentioned (C) solvent. Moreover, when the constituent of this invention is excellent in storage stability, for example, change of the spreading thickness before and behind one-month neglect is measured at 40 degrees C among glass well-closed containers, the rate of change is less than 10%.

[0046] In order to form a film using the constituent of this invention, the constituent of this invention is first applied to a substrate, and a paint film is formed. Here, as a substrate which can apply the constituent of this invention, a semiconductor, glass, Ceramics Sub-Division, a metal, etc. are mentioned. Moreover, a spin coat, dipping, a roller blade, etc. are mentioned as the spreading method. Especially the constituents of this invention are a silicon wafer and SiO₂. It is applied on a wafer, a SiN wafer, etc. and is suitable for considering it as an insulating film.

[0047] The thickness in this case can form a paint film about 0.1-3 micrometers thick by

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coating once as desiccation thickness by about 0.05-1.5 micrometers in thickness, and 2 times coating. The thickness of the paint film to form is usually 0.2-20 micrometers. As the heating method in this case, a hot plate, oven, Furness, etc. can be used and it can carry out under nitrogen-gas-atmosphere mind, argon atmosphere, and a vacuum under the air as a heating atmosphere under the reduced pressure which controlled oxygen concentration etc. As this heating method, heat the formed paint film at the temperature of under the decomposition temperature of the ** above-mentioned (B) component, and a part of (A) component is stiffened. Subsequently, it heats from the temperature more than the decomposition temperature of the above-mentioned (B) component to final cure temperature, and the method of using as the hardened material of a low density, the method of heating, making harden at the temperature more than the decomposition temperature of the ** above-mentioned (B) component, and using as the hardened material of a low density, etc. are mentioned.

[0048] Moreover, since the cure rate of the above-mentioned (A) component and the catabolic rate of the (B) component are controlled, if needed, it can heat gradually or atmosphere, such as nitrogen, air, oxygen, and a reduced pressure, can be chosen. Usually, since 200-400 degrees C of decomposition temperature of the (B) component is usually 200-350 degrees C preferably, finally a paint film includes the process heated beyond this temperature. As for this process, it is desirable to be carried out under a reduced pressure state or inert gas.

[0049] thus, the low density-ized film of this invention obtained -- a film density -- usually -- 0.35-1.2g/cm³ -- desirable -- 0.4-1.1g/cm³ -- further -- desirable -- 0.5-1.0g/cm³ it is . 0.35g/cm³ The mechanical strength of a paint film falls in the following, and, on the other hand, it is 1.2g/cm³. Low permittivity will not be obtained if it exceeds. The content of the (B) component in the constituent for film formation in this invention can adjust adjustment of this film density easily.

[0050] Furthermore, when the low density-ized film of this invention has the feature at the point that absorptivity is low, for example, a paint film is neglected by the environment of 127-degree-C, 2.5atm, and 100%RH** for 1 hour, the adsorption of water to a paint film is not accepted from IR-spectrum observation of the paint film after neglect. This absorptivity can be attained by considering it as the range given [the content of the component (A-3) in the constituent for film formation in this invention] in this invention.

[0051] furthermore -- the permittivity of the low density-ized film of this invention is low permittivity -- usually -- 2.6-1.2 -- desirable -- 2.5-1.2 -- it is 2.4-1.2 still more preferably. The content of the (B) component in the constituent for film formation of this invention can adjust this permittivity.

[0052] The low density-ized film of this invention has the feature at the point whose thickness homogeneity is very good. Use the constituent for this film formation on a 8-inch silicon wafer, and a spin coater For example, after spreading, after heating for 5 minutes at 200 degrees C

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under nitrogen subsequently for 5 minutes at 80 degrees C among the air, it heats every [during 30 minutes] under a vacuum further, respectively in order (340 degrees C, 360 degrees C, and 380 degrees C), it heats at 425 degrees C under a vacuum further for 1 hour, and a paint film is formed. the thickness of the obtained film -- an optical thickness gage (the product made by Rudolph Technologies --) Spectra When 50 points are measured in a film surface using Laser200, 3sigma (sigma: standard deviation) is less than 2%, and thickness homogeneity is good and very desirable as an interlayer insulation film material between detailed wiring. This very good thickness homogeneity can be attained by using the (B) component in this invention.

[0053] The low density film of this invention excels [low density film] in insulation, and The homogeneity of a spreading film, permittivity characteristics, LSI, a system LSI since it excels in the surface hardness of the crack-proof nature of a paint film, and a paint film, It is useful for the use of overcoats, such as interlayer insulation films for semiconductor devices, such as DRAM, SDRAM, RDRAM, and D-RDRAM, and a surface coat film of a semiconductor device, the interlayer insulation film of a multilevel interconnection board, the overcoat for liquid-crystal-display elements, an insulating prevention film, etc., etc.

[0054]

[Example] Hereafter, an example is given and this invention is explained still more concretely. However, the following descriptions show the example of a mode of this invention generally, a Reason in particular does not have them, and this invention is not limited by this description. Moreover, the part in an example and a comparative example and % show that it is weight part and weight %, respectively, unless it mentions specially.

[0055] Preparation of an example 1 (A) component; tetramethoxy gardenia fruit run 101.3g (complete-hydrolysis condensate conversion: 40.0g), Methyl trimetoxysilane 203.0g (complete-hydrolysis condensate conversion: 100.0g), Dimethyldimethoxysilane 97.3g (complete-hydrolysis condensate conversion: 60.0g), [propylene glycol monopropyl ether 559.3g and a methyl n-pentyl ketone / 239.7g / mixed solution] 1.0g of maleic acid -- [-- total mole [of the R1 O group of a catalyst / compound (1) - (3)] = -- a 0.001mol ratio -- the R1 O group of H2 O / 157.7g of water [compound (1) - (3) -- the total -- the aqueous solution melted to mole =1.0mol ratio] was dropped over 1 hour at the room temperature. condensing after [after the end of dropping of a mixture, and] making it react at 60 more degrees C for 2 hours until it becomes the total amount of solutions of 1,000g under a reduced pressure -- polysiloxane of 20% of a solid content -- sol was obtained.

constituent preparation-; -- the polysiloxane obtained above -- the mixture which added polyethylene glycol mono-undecanoic acid ester (the number of oxyethylene repetitions = 10) 3.5g to Sol 100g (20g of solid content), and was obtained -- a 8-inch silicon wafer top -- a spin coat method after applying and heating for 5 minutes at 200 degrees C under nitrogen

subsequently for 5 minutes at 80 degrees C among the air, it heated every [during 30 minutes] under the vacuum further, respectively in order (340 degrees C, 360 degrees C, and 380 degrees C), and heated at 425 degrees C under the vacuum further for 1 hour, and the water-white film was formed. Furthermore, the constituent and film which were obtained were evaluated as follows. A result is shown in Table 1.

[0056] 80ml of constituents for film formation in evaluation 1. storage stability this invention of the constituent for film formation were put into glass screw-cap bottles with a capacity of 100ml, and were sealed, and it was neglected for one month in the 40-degree-C autoclave. When the sample before and behind neglect is applied on 2,500rpm and the spin coat conditions for 31 seconds and it calcinates by the method of the example 1 above-mentioned description ***** -- an optical thickness gage (the product made by Rudolph Technologies --) Spectra It measured by Laser200, the rate of change $\{[(\text{thickness of sample before neglect} - \text{thickness of sample after neglect}) / \text{thickness of sample before neglect}] \times 100\}$ was computed, and the following basis estimated.

O : thickness rate-of-change $< 10\%$ x : thickness rate-of-change $\geq 10\%$ [0057] 2. On the 8-inch silicon wafer, the spin coater was used and the constituent for thickness homogeneity film formation was applied on the number of rotations of 2,500rpm, and the conditions for 31 seconds. Then, 50 thickness when calcinating by the method of the example 1 above-mentioned description was measured in the film surface using the optical thickness gage (the product made by Rudolph Technologies, Spectra Laser200). 3sigma (sigma: standard deviation) of the obtained thickness was calculated, and the following basis estimated.

O : -- less than $[3\sigma 2\%]$ x: of a paint film -- $3\sigma 2\%$ or more of a paint film [0058] 3. the aluminum electrode pattern was made to form with vacuum deposition to a permittivity profit **** film, and the sample for permittivity measurement was created. On the frequency with a frequency of 100kHz, the permittivity of the paint film concerned was measured for this sample by the CV method using YOKOGAWA and the Hewlett Packard Co. make, a HP16451B electrode, and HP4284A precision LCR meter. A result is shown in Table 1.

[0059] 4. The film density density was computed from membranous thickness, the volume for which it asked from a membranous area, and membranous weight.

5. the elastic-modulus profit **** film was measured with the continuation rigidity measuring method using the nano indenter XP (made by a nano instrument company).

[0060] 6. the absorptivity profit **** paint film was neglected by the environment of 127-degree-C, 2.5atm, and 100%RH** for 1 hour, and the IR spectrum of the paint film after neglect was observed. As compared with the IR spectrum of the paint film before neglect, the existence of absorption resulting from H₂O of the 3,500cm⁻¹ neighborhood was observed, and absorptivity was evaluated in accordance with the following basis.

O : those with absorption-less x:absorption [0061] Preparation of an example 2 (A) component;

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tetramethoxy gardenia fruit run 152.0g (complete-hydrolysis condensate conversion: 60.0g), Methyl trimetoxysilane 284.1g (complete-hydrolysis condensate conversion: 140.0g), [a propylene glycol monomethyl ether / 798.8g / mixed solution] the R1 O group of a catalyst / 1.0g of oxalic acid [compound (2) - (3) -- the total -- mole =0.001mol ratio] -- the R1 O group of H2 O / 157.7g of water [compound (2) - (3) -- the total -- the aqueous solution melted to mole =1.0mol ratio] was dropped over 1 hour at the room temperature. condensing after [after the end of dropping of a mixture, and] making it react at 60 more degrees C for 2 hours until it becomes the total amount of solutions of 1,000g under a reduced pressure -- polysiloxane of 20% of a solid content -- sol was obtained.

constituent preparation-; -- the polysiloxane obtained above -- the mixture which added polyethylene glycol mono-undecanoic acid ester (the number of oxyethylene repetitions = 10) 8.6g to Sol 100g (20g of solid content), and was obtained -- a 8-inch silicon wafer top -- a spin coat method after applying and heating for 5 minutes at 200 degrees C under nitrogen subsequently for 5 minutes at 80 degrees C among the air, it heated every [during 30 minutes] under the vacuum further, respectively in order (340 degrees C, 360 degrees C, and 380 degrees C), and heated at 425 degrees C under the vacuum further for 1 hour, and the water-white film was formed. Evaluation of the constituent and film which were obtained was performed like the example 1. A result is shown in Table 1.

[0062] Preparation of an example 3 (A) component; tetramethoxy gardenia fruit run 101.3g (complete-hydrolysis condensate conversion: 40.0g), Methyl trimetoxysilane 324.7g (complete-hydrolysis condensate conversion: 160.0g), [the mixed solution of propylene glycol monopropyl ether 783.3g and G i-propoxy bis(ethyl acetoacetate) titanium [total mole =0.004mol ratio of the R1 O group of a catalyst / compound (2) - (3)]] the R1 O group of H2 O / 176.8g of water [compound (2) - (3) -- the total -- mole =1.0mol ratio] -- 60 degrees C -- warming -- it was dropped over 1 hour in the bottom. condensing until it adds Acetylacetone 100.0g and becomes the total amount of solutions of 1,000g under a reduced pressure after that, after making it react at 60 more degrees C for 2 hours, after the end of dropping of a mixture, -- polysiloxane of 20% of a solid content -- sol was obtained.

constituent preparation-; -- the polysiloxane obtained above -- the mixture which added polyethylene glycol mono-undecanoic acid ester (the number of oxyethylene repetitions = 10) 3.5g to Sol 100g (20g of solid content), and was obtained -- a 8-inch silicon wafer top -- a spin coat method after applying and heating for 5 minutes at 200 degrees C under nitrogen subsequently for 5 minutes at 80 degrees C among the air, it heated every [during 30 minutes] under the vacuum further, respectively in order (340 degrees C, 360 degrees C, and 380 degrees C), and heated at 425 degrees C under the vacuum further for 1 hour, and the water-white film was formed. Evaluation of the constituent and film which were obtained was performed like the example 1. A result is shown in Table 1.

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[0063] In comparative example 1 example 1, except not using polyethylene glycol mono-undecanoic acid ester (the number of oxyethylene repetitions = 10), the constituent was manufactured like the example 1, and it applied to the substrate, it heated, and the film was formed. Evaluation of the obtained film was performed like the example 1. A result is shown in Table 1.

[0064] Set in the comparative example 2 example 1. Except not using propylene glycol monopropyl ether 559.3g and methyl n-pentyl ketone 239.7g, but using Tetrahydrofuran 799.0g instead, the constituent was manufactured like the example 1, and it applied to the substrate, it heated, and the film was formed. Evaluation of the constituent and film which were obtained was performed like the example 1. A result is shown in Table 1.

[0065] Set in the comparative example 3 example 1. Polyethylene glycol mono-undecanoic acid ester (the number of oxyethylene repetitions = 10) 3.5g is not used. Except using 3.5g of polymethyl methacrylate resin (molecular weight 20,000) instead, the constituent was manufactured like the example 1, and it applied to the substrate, it heated, and the film was formed. Evaluation of the constituent and film which were obtained was performed like the example 1. A result is shown in Table 1.

[0066]

[Table 1]

	実施例			比較例		
	1	2	3	1	2	3
貯蔵安定性	○	○	○	○	×	○
誘電率	2.4	2.3	2.4	2.8	2.5	2.5
膜密度 (g/cm ³)	1.2	1.1	1.2	1.4	1.2	1.3
塗膜均一性	○	○	○	○	○	×
弾性率 (GPa)	3.9	3.3	3.4	5.5	3.6	3.2
吸水性	○	○	○	○	○	○

[0067]

[Effect of the Invention] the film which according to this invention is excellent in storage stability and is obtained -- homogeneity and a low density -- low permittivity -- low -- the absorptivity

constituent for film formation can be offered.

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[Translation done.]